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硕　士　学　位　论　文

基于拓扑绝缘体薄膜的 3D 超晶格纳米结构热电性能的几何依赖性与优化分析

Theoretical Study on the Geometrical Size Dependence and Optimization Analysis of Thermoelectric Performance for Topological Insulator Film Based Three-dimensional Superlattice Nanostructures

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摘　要

热电器件作为利用塞贝克效应能使废热产生电能的固体电子器件，是解决可持续发展中能源问题的重要解决方案。 但迄今为止，热电材料普遍较低的热电转换效率限制了该功能器件在温差发电的应用前景，使得寻找足够高热电 ZT 值的热电材料的理论研究和实验研究近几十年来备受关注。事实上，最近人们分别在 $AgPb_{18}SbTe_{20}$，Bi_2Te_3 基材料的超晶格形态，填充 Skutterudite 型材料，$NaCo_2O_4$, $Ca_3Co_4O_9$ 等金属氧化物材料均测量到了较高的热电 ZT 值，也总结出了提升热电材料 ZT 值的基本手段，即使得材料具有较低的晶格热导率与电导率，同时拥有较大的塞贝克系数和较高的电导率。而作为广泛用于热电制冷的 Bi_2Te_3 型热电材料，如何用低维超晶格化的方法高效提升其 ZT 值一直是个理论和实验研究热点。

最近，随着具有奇特物理性质的三维拓扑绝缘体之发现，以及常见的 Bi_2Te_3 热电材料恰好也可以是拓扑绝缘体，引起了人们利用拓扑绝缘体拥有高迁移率表面态与较窄的体带隙的独特性质来设计具有更高热电优值材料的广泛兴趣。 研究者期望获得相比块体 Bi_2Te_3 基(ZT～1)热电材料显著提升的 ZT 值，以开发下一代高效热电器件。

本文基于 Zheyong.Fan 等人的设计的基于三维拓扑绝缘体 Bi_2Te_3 薄膜的 3D 纳米超晶格结构来提升热电 ZT 值的理论研究。 该 3D 超晶格结构利用真空孔洞的对声子界面散射降低了晶格热导率，同时利用表面态电子的高迁移率，以及界面散射的能量滤波作用散射低能量电子，于是在一定化学势和温度下获得了很高的热电 ZT 值。我们利用修正有效近似理论和 Landauer 输运理论分别数值计算了体系热电性质与其几个关键几何参数之间的依赖关系，详细描述其随几何参数增大而演化的模式并用基本输运理论解释其中机制，分析热电 ZT 值对几何参数组合的优化， 探寻其中的特征几何尺寸是如何通过影响电子输运和晶格热输运而改变该 3D 超晶格结果的热电转换效率。本文工作的意义在于以开辟通过纳米几何优化设计的方法来提高低维超晶格化的拓扑绝缘体热电材料之 ZT 值的新途径，同时为在其他低维材料中研究热电性能的尺寸依赖性提供参考。

我们将在本文第一，二章介绍热电效应基本原理，热电 ZT 值提升理论和热电材料研究的最新进展，以及电子输运系数和晶格热导率的基本计算理论。 在

第三章详细介绍本文研究的背景与采用的计算方法。将第四，五，六，七章作为数值计算结果的讨论分析章节，分别详细阐述了得到的数值结果: 1）3D 超晶格热电输运对几何放大倍数 m 的标度关系 2）晶格热导率对 3D 超晶格几何参数的依赖及敏感度分析 3）3D 超晶格结构的电导率，塞贝克系数，功率因子，电子的热导率贡献对几何参数的依赖性与敏感度分析 4）热电 ZT 优值对该 3D 超晶格结构几何参数的依赖性与敏感度分析及其优化结果。

我们将在第八章作全文总结，并探讨本课题研究对于实验上通过调节几何尺寸来优化该 3D 超晶格结构的热电 ZT 值的指导意义。

本论文数值计算与绘图均采用 Mathematica^{R} 作为科学计算编程语言。

关 键 词：热电 ZT 优值 几何参数优化 塞贝克系数 拓扑绝缘体超晶格 有效介质近似

Abstract

Taking advantage of the Seebeck effect, thermoelectric devices are the solid state electronic devices using waste heat to generate electricity, making itself an important solution for solving the energy problems to achieve sustainable development. However, so far the thermoelectric conversion efficiency of thermoelectric materials is generally a limitation of this type of functional devices in the prospect thermoelectric power generation, making people concerned about the theoretical and experimental research in recent decades to find a sufficiently high ZT value of thermoelectric thermoelectric materials. In fact, recently people already measured thermoelectric higher ZT values in $AgPb_{18}SbTe_{20}$, superlattices form of Bi_2Te_3-based materials, filling Skutterudite material, $NaCo_2O_4$, $Ca_3Co_4O_9$ other metal oxide materials, and also gived a summary of the basic approaches to enhance the ZT values of thermoelectric materials, namely trying to achieve a low lattice thermal conductivity and electric thermal conductivity, with a large Seebeck coefficient and high electrical conductivity. As the Bi_2Te_3-type thermoelectric material widely used in thermoelectric cooling, and how to use low-dimensional superlattice of methods to enhance the efficiency of its ZT figure of merit has always been a theoretical and experimental research focus.

Recently, with the discovery of three-dimensional topological insulator with novel physical properties, as well as common thermoelectric materials Bi_2Te_3 can also be a strong topological insulators, people were widely attracted by the idea of applying the unique nature oftopological insulator that a surface state with high mobility and a narrow bulk gap, to design thermoelectric material with a much higher figure of merit. The researchers expect to obtain a significantly improved thermoelectric ZT value compared to the bulk Bi_2Te_3 material (ZT~1) , aimed at developing the next generation of highly efficient thermoelectric devices.

This Master's thesis is based on a research by Zheyong.Fan on three-dimensional topological insulators Bi_2Te_3 film constructed superlattice structure design, which is

aimed to gain an enhanced ZT figure of merit of this 3D thermoelectric nanostructure through theoretical study.

Taking advantage of the phonon scattering effect with the vacuum holes in the superlattice structure, it greately reduces the lattice thermal conductivity, and at the same time by applying the high electron mobility of the surface state, and the energy filtering of the interface scattering of low energy electrons scattered, and so in a certain combination of chemical potential and temperature, this 3D superlattice obtain a high ZT value.

We use the modified effective meidum approximation theory and Landauer transport theory to perform numerical calculations of the dependence of thermoelectric properties of the system with its several key geometric parameters, described in detail its evolution pattern with the geometric parameters increases and explain the inner mechanism by the fundamental transport theory, analyzed the optimization of thermoelectric ZT values with its geometric parameters combination, discussed and explored how the characteristic geometry changes the results of the 3D superlattice thermoelectric conversion efficiency by affecting electron transport and the lattice thermal transport.

The significance of this work is to open up a new approach based on nano-geometry optimization methods through which we can improve the ZT figure of merit in low-dimensional thermoelectric materials of 3D topological insulator superlattice o, as well as providing theoretical reference for studies on the size-dependence thermoelectric properties of other low-dimensional materials and their geometrical optimization design.

The following are what we will study in this thesis.

In the first two chapters, we will introduce the basic principles of thermoelectric effect, and basic calculation theory of electron transport coefficients and the lattice thermal conductivity and research progress thermoelectric materials study,disscuss the latest theories on enhancement of thermoelectric ZT values. Detailed background of the main study of this thesisi and calculation methods used will be given in the third chapter.

As the numerical results of the discussion and analysis section, the fourth, fifth, six, seven chapters elaborated on the numerical results obtained, respectively on 1) the scaling relation of 3D superlattice thermoelectric transport on geometric magnification m. 2) dependence on the lattice thermal conductivity of the superlattice 3D geometric parameters and relevant sensitivity analysis. 3) the dependence of the electronic conductivity, the Seebeck coefficient, the power factor, the thermal conductivity contribution of electrons with of the 3D superlattice structure, and the sensitivity analysis of geometric parameters. 4) dependence of thermoelectric figure of merit ZT on 3D superlattice geometric parameters. and sensitivity analysis , and also the optimization of the results of ZT.

We will make a full summary in Chapter 8, and explore the experimental guidance. of this research to optimize the ZT values of 3D thermoelectric superlattice structure by adjusting its geometry.

In this paper, all the numerical calculations and graphing are programmed using Mathematica^{R} as the scientific computing programming language.

Keywords: ZT Figure of Merit , Geometrical Parameters Optimization, Seebeck coefficient, Topological Insulator Based Superlattice, Effective Medium Approximation.

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